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Impacts of climate change on regional and urban air quality in the eastern United States: Role of meteorology

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[1] The effects of climate change on ozone and $PM_{2.5}$ concentrations over the eastern United States were investigated using the Global-Regional Coupled Air Pollution modeling System (GRE-CAPS). GRE-CAPS consists of the Goddard Institute for Space Studies (GISS) II' general circulation model with aerosol processes and ozone chemistry, the fifth-generation PSU/NCAR mesoscale model (MM5) regional meteorological model, and the Comprehensive Air Quality Model with Extensions (CAMx) with aerosol (PM) processes developed at Carnegie Mellon University (PMCAMx) regional chemical transport model. A set of five present-day Januaries and six present-day Julys was simulated using GRE-CAPS. The present-day model predictions (2000s) were compared to model predictions for a set of five future Januaries and Julys. The future time period investigated was the 2050s, using the Intergovernmental Panel on Climate Change A2 scenario. U.S. emissions of biogenic and anthropogenic precursors were held constant so that the effects of climate change alone could be calculated. Climate change led to a decrease in U.S. land cell average January $PM_{2.5}$ concentrations of 0.3 μg m^{-3} and an increase of July $PM_{2.5}$ of 2.5 μg m^{-3}. The changes in PM in the Northeast were of the opposite sign of the domain-wide averages. The response in January was due largely to increased precipitation, while the response in July was due primarily to decreased ventilation, as indicated by decreases in mixing height and wind speed, with increases in sulfate being the largest response by a single species. The U.S. land cell average change in July daily maximum 8-h ozone concentration was +1.7 ppb, though the increases in cities in the Southeast were up to 15 ppb. In spite of the large differences in ozone in many areas, the changes in ozone concentration were not statistically significant over most of the domain because of large interannual variability. In separate simulations to test the sensitivity of ozone concentrations to biogenic emissions, a 25% increase in biogenic U.S. volatile organic compound emissions led to an additional increase in land cell average ozone of 0.7 ppb, though the increased ozone resulting from increased biogenics was largely statistically insignificant.

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1. Introduction

[2] High concentrations of ozone (O_3) and particulate matter (PM) are two persistent problems that affect air quality. Ozone and PM have been shown to have effects on human health [*Bernard et al.*, 2001; *Levy et al.*, 2001; *Schwartz et al.*, 1996] and on climate [*Intergovernmental Panel on Climate Change* (IPCC), 2001]. PM also impacts visibility, while ozone causes crop damage [*Heck et al.*, 1982].

[3] Previous work has shown that concentrations of ozone and PM are sensitive to changes in meteorology. Modeling studies [*Baertsch-Ritter et al.*, 2004; *Dawson et al.*, 2007a, 2007b] have examined these sensitivities to a suite of meteorological variables. Ozone concentrations are rather sensitive to temperature, humidity, wind speed, and mixing height, while PM concentrations are sensitive to the same meteorological variables plus precipitation. Changes in climate over the next century are expected to result in changes in many or all of these meteorological parameters, which could have important impacts on air quality.

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[4] To investigate the effects of global climate change on regional and urban air quality in the eastern United States, the Global-Regional Coupled Air Pollution modeling System (GRE-CAPS) was developed [*Dawson et al.*, 2008]. GRE-CAPS links the Goddard Institute for Space Studies (GISS) II' GCM with aerosol processes and ozone chemistry (GISS II' is a chemistry-climate model, CCM) [*Hansen et al.*, 1983; *Mickley et al.*, 1999; *Adams et al.*, 1999; *Liao et al.*, 2003], the fifth-generation PSU/NCAR mesoscale model (MM5) regional meteorological model [*Grell et al.*, 1994], and the Comprehensive Air Quality Model with Extensions (CAMx) with aerosol (PM) processes developed at Carnegie Mellon University (PMCAMx) regional CTM [*Gaydos et al.*, 2007; *Karydis et al.*, 2007]. The GRE-CAPS system was shown to reproduce present-day ozone and $PM_{2.5}$ concentrations (total and individual species) with similar accuracy to that of the standard configuration of PMCAMx (driven by assimilated meteorology), in spite of the use of GCM-generated meteorology in GRE-CAPS [*Dawson et al.*, 2008].

[5] Other researchers have used similar modeling systems to examine the effects of global change on regional air quality. *Hogrefe et al.* [2004a, 2004b] used meteorology generated by the GISS GCM [*Russell et al.*, 1995] and downscaled by MM5 to simulate present-day and future climates in a regional CTM (CMAQ) [*Byun and Ching*, 1999]. The purpose of the work of *Hogrefe et al.* [2004a, 2004b] was to simulate present-day and future ozone concentrations over the eastern USA, using the *IPCC* [2000] A2 scenario for the future. This modeling system predicted an increase in summertime daily maximum 8-h average (MDA8) O_3 in the 2050s compared to the present-day values of 4.2 ppb due to climate change and changes in climate-sensitive biogenic emissions [*Hogrefe et al.*, 2004b]. That work concluded that changes in climate alone could have an appreciable effect on future ozone concentrations. *Tagaris et al.* [2007] also used downscaled meteorology to simulate ozone and $PM_{2.5}$ over the United States in the present day and in 2050, using the IPCC A1B scenario for the future. They predicted a rather small impact of climate on pollutant concentrations, with predicted emissions changes having a much larger impact. The effects of climate alone on air quality included a small change in ozone concentrations in the summer and decreases in summer $PM_{2.5}$ due to increased precipitation. The differences in predicted effects of climate among various studies are likely due to differences in chosen scenarios and predicted meteorological changes.

[6] Additional work has focused on predicting the effects of climate change on air quality using global-scale models with no downscaling. *Racherla and Adams* [2006] and *Murazaki and Hess* [2006] predicted increases in ozone over the eastern United States for the A2 and A1 scenarios respectively. *Racherla and Adams* [2006] also linked changes in global PM concentrations largely to changes in precipitation; they calculated an increase in PM in June and July due to decreased precipitation.

[7] The discrepancies among model predictions indicate that there is still significant uncertainty in the expected effect of climate on ozone concentrations. The effect of climate on PM concentrations is even more uncertain than the effect on ozone concentrations. This work provides an estimate for the changes in $PM_{2.5}$ and revisits the predicted changes in ozone. The GRE-CAPS system [*Dawson et al.*, 2008] is used to simulate the present-day and the 2050s climates and examine the effect of changes in climate on ozone and $PM_{2.5}$ concentrations. GRE-CAPS has shown improved present-day model performance compared to other coupled modeling systems (i.e., smaller biases in O_3 and $PM_{2.5}$ predictions) [*Dawson et al.*, 2008].

2. Methods

[8] The model-predicted present-day ozone and $PM_{2.5}$ concentrations were compared to model-predicted future concentrations using the GRE-CAPS modeling system. The same six Julys and five Januaries simulated by *Dawson et al.* [2008] were used to represent the present day in this comparison. The future global meteorology was generated by the GISS II' GCM and was based on the *IPCC* [2000] A2 scenario for the 2050s [*Racherla and Adams*, 2006]. Previous studies, such as that of *Cess et al.* [1990], have put the GISS GCM II' at the higher end of GCM-predicted climate sensitivities. Much of the IPCC interscenario divergence in climate occurs after 2050, so this model-predicted future climate can be thought of as an estimate for other scenarios (such as the B1 scenario) as well.

[9] The GRE-CAPS system, described more fully by *Dawson et al.* [2008], consists of three component models: the GISS II' GCM with online chemistry, the MM5 regional meteorological model [*Grell et al.*, 1994], and the PMCAMx regional CTM [*Gaydos et al.*, 2007; *Karydis et al.*, 2007]. Climate and chemical concentration fields were generated for present-day and 2050s conditions at the global scale, $4^\circ \times 5^\circ$, by GISS II'. The GCM was run using present-day anthropogenic emissions and climate-sensitive biogenic emissions. Both the present and future climate predictions in the GCM were driven by ocean boundary conditions as described by *Racherla and Adams* [2006] The meteorology generated by GISS II' was downscaled by providing boundary conditions, updated every 4 h, to MM5, which was run, without periodic reinitialization, at a spatial scale of 100 km $\times$ 100 km over the entire United States and a nested grid of 36 km $\times$ 36 km over the eastern United States. This downscaled meteorology was used in a PMCAMx simulation, using the CB-IV gas phase mechanism [*Gery et al.*, 1989] and the aerosol modules summarized by *Gaydos et al.* [2007], at 36 km $\times$ 36 km, over the eastern United States. The emissions inventory used was the Midwest Regional Planning Organization's Base E inventory [*Lake Michigan Air Directors Consortium*, 2003], including BIOME3 biogenics [*Wilkinson and Janssen*, 2001]. All emissions, including biogenics, were for July 2001 or January 2002 (except in section 3.4, in which biogenic emissions were increased by 25%). The chemical concentrations predicted by the GCM around the outside of the PMCAMx modeling domain were used as boundary conditions, updated every 4 h. This coupled modeling system was evaluated by *Dawson et al.* [2008]; GRE-CAPS was found to have biases and errors similar to those of standard PMCAMx, in spite of GRE-CAPS's use of GCM-generated, nonassimilated meteorology. The future climate was represented by five Julys and Januaries in GRE-CAPS.

Table 1. Regional Changes (Future – Present) in Model-Predicted Meteorological Parameters[a]

	Plains	Midwest	Northeast	TX/OK	Southeast
		January			
Surface T	−0.3 K	+0.7 K	+1.9 K	−2.5 K	−0.3 K
Column average T	−0.4 K	0.0 K	+1.6 K	−0.9 K	+0.4 K
Precipitation	+85%	+63%	*+73%*	+2%	+26%
Surface wind speed	+11%	+7%	−7%	−8%	−2%
Surface absolute humidity	−1%	+3%	+11%	−8%	+1%
Mixing height	+5%	*+23%*	+4%	−22%	+3%
		July			
Surface T	+1.1 K	+1.2 K	+1.0 K	−0.4 K	+0.9 K
Column average T	+1.4 K	+1.8 K	+1.0 K	+0.6 K	+2.3 K
Precipitation	+22%	+13%	*−40%*	−26%	*+88%*
Surface wind speed	−3%	−13%	−14%	−27%	−19%
Surface absolute humidity	+14%	+13%	−1%	+21%	+31%
Mixing height	−10%	−16%	−4%	−36%	−38%

[a]Changes in italics were marginally statistically significant, with *t* test *p* values between 0.10 and 0.05. Other changes were not statistically significant.

The present and future global climate and chemistry simulations were described by *Racherla and Adams* [2006].

[10] Emissions, both anthropogenic and biogenic, were held at present-day levels in the regional CTM (PMCAMx) so that the meteorological effects of climate alone could be analyzed, though biogenic emissions varied with climate at the global scale. The effects of anthropogenic emissions changes will be examined in future work. The effects of biogenic emissions changes over the United States are discussed in section 3.4 later in this paper. For ozone, changes in July–average daily peak and MDA8 concentrations were examined as was the number of grid cells exceeding the air quality standard of an 8-h average concentration over 0.08 ppm (84 ppb). Present and future climate concentrations of total $PM_{2.5}$ as well as $PM_{2.5}$ sulfate, nitrate, ammonium, and organics were compared. Ozone concentrations were considered only in July, while $PM_{2.5}$ concentrations were investigated for both January and July.

[11] Chemical boundary conditions were based on the species concentrations predicted by GISS II' around the outside of the eastern U.S. domain, updated every 4 h, so that transport changes due to changes in meteorology could also be taken into account. The simulations of future (2050s) climate used the same chemical boundary conditions (representing long-distance transport) around the eastern United States as did the simulations of present-day climate. This allows the isolation of regional climate effects from global-scale transport effects, but it may lead to inconsistencies between the chemical boundary conditions and the hourly meteorology. Because of the unreasonably large amounts of dust and sea salt entering the eastern U.S. modeling domain from the GISS II'-generated boundary conditions [*Dawson et al.*, 2008], only sulfate, nitrate, ammonium, and organic (both primary and secondary) PM are included in the results of this study.

[12] Five additional Julys with future climate were simulated in PMCAMx, with U.S. emissions of isoprene and biogenic olefins were increased by 25% to examine the effect of changes in climate-sensitive biogenic volatile organic compounds (VOCs) on ozone and PM concentrations. The 25% increase in biogenics is based on *Racherla and Adams* [2006], in which increases between 20 and 30% in biogenics were predicted for the same scenario.

3. Results and Discussion

3.1. Meteorology

[13] The predicted regional changes in several meteorological variables between the present and future are shown in Table 1 for the regions defined in Figure 1. In January, precipitation changes appear to be the dominant meteorological change in most of the domain. Large increases in precipitation would presumably lead to increased removal and reduced concentrations of $PM_{2.5}$. In July, there is no clearly dominant effect, though both mixing height and wind speed decreased over most of the domain. The changes in July mixing height and wind speed around the eastern United States are shown in Figure 1. Most of the July

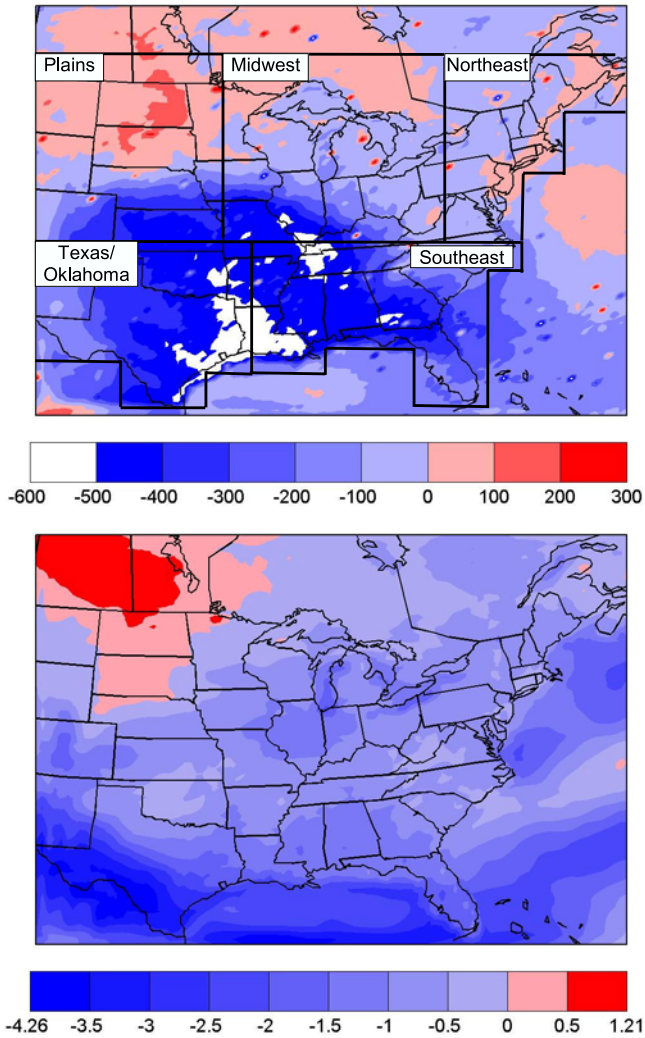

Figure 1. Changes (future – present) in average (top) mixing height (m) and (bottom) wind speed (m s^{-1}) in July. The locations of five subregions used for analysis are also shown. (Statistical significance information is noted in Table 1.)

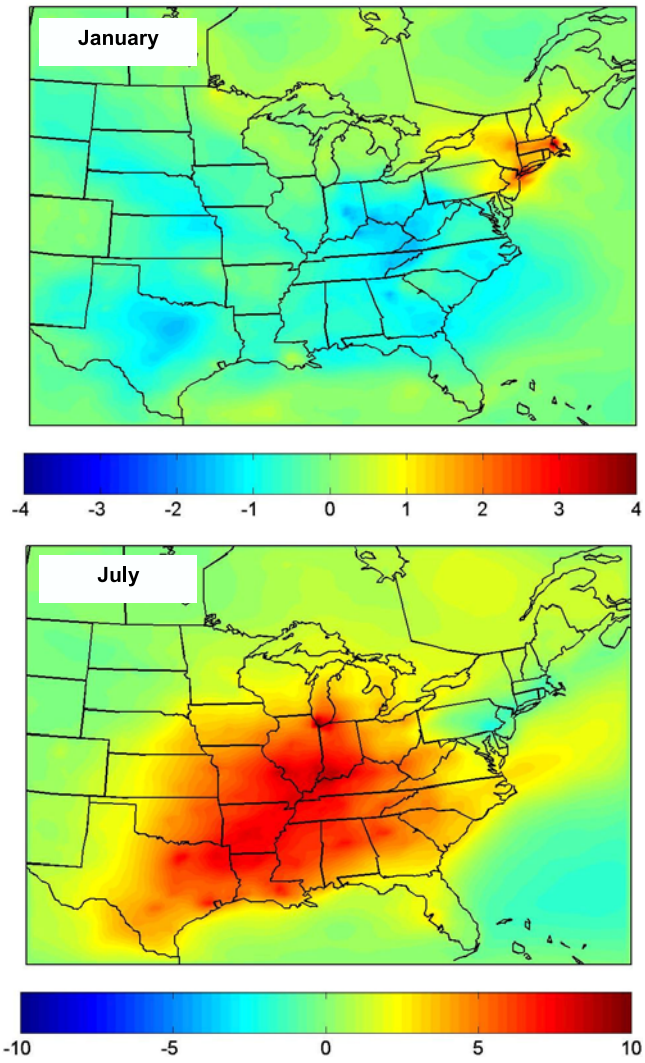

Figure 2. Changes in average (top) January and (bottom) July $PM_{2.5}$ concentrations ($\mu g\ m^{-3}$) between future climate and present climate ensembles.

meteorological changes in Table 1, especially decreased wind speed and mixing height and increased temperature, would result in increased $PM_{2.5}$ concentrations according to the results from *Dawson et al.* [2007b]. In the work by *Dawson et al.* [2007a], decreases in mixing height and wind speed and increases in temperature generally led to increased domain average ozone concentrations, while increased absolute humidity led to smaller and more complex effects. Because there is not a statistically significant difference in most of the quantities in Table 1, it cannot be definitively established that the differences between the present and future simulations are not simply due to interannual variability.

[14] These changes are largely consistent with those predicted in other work. The results for ventilation are similar to those of *Leung and Gustafson* [2005], who predicted decreased summertime mixing heights and wind speeds over much of the South and Midwest for the 2050s and the IPCC A1B scenario using dynamic downscaling. *Mickley et al.* [2004], however, calculated increases in mixing heights of 100–240 m over the Northeast and Midwest between the present-day and the 2050s, though this study used only a GCM with relatively coarse vertical resolution. *Mickley et al.* [2004] did find an increase in stagnation over the eastern United States and, therefore, decreased wind speeds. *Murazaki and Hess* [2006] noted "more sluggish circulation" under a future (2090s, IPCC A1 scenario) climate, indicating decreased wind speeds, though no significant changes in mixing heights were reported.

[15] The changes in temperature resulting from downscaling (Table 1) are generally somewhat smaller than those predicted by the GCM itself, in which average eastern U.S. summer surface level temperature increased by 1.6 K and annual average temperature increased by 1.7 K [*Racherla and Adams*, 2006]. *Leung and Gustafson* [2005] predicted similarly small warming compared to Table 1 between the present and the 2050s over the Midwest, Southeast, and Northeast during the summer and average warming between 0 and 3 K in the winter for the A1B scenario. The summertime increases in temperature predicted by *Murazaki and Hess* [2006] for the A1 scenario for the 2090s ranged from approximately 1.5 to 3.5 K, resulting in a larger average increase than that predicted by GRE-CAPS. The downscaled 2050s summer temperature increase predicted by *Hogrefe et al.* [2004b] was 2.7 K, compared to a smaller increase of 1.9 K predicted by the GCM. In their predictions of 2080s climate using an ensemble of GCMs, *Giorgi and Mearns* [2002] calculated a wide range of predicted changes in temperature (increases between 2 and 7 K) and precipitation (−25% to +30%) for the A2 scenario. Over eastern North America, GCMs predicted small precipitation changes in both seasons. In their intermodel comparison of predicted changes in temperature and precipitation, *IPCC* [2007] noted that GCMs generally predict greater-than-global-average warming in both winter and summer in eastern North America for the A2 and B2 scenarios. Additionally, the *IPCC* [2001] analysis concluded that GCMs generally predict increased precipitation over much of eastern North America in the winter, though there was no intermodel consensus for summer precipitation changes.

[16] There were several significant changes in the GRE-CAPS-predicted interannual variability of the area-weighted averages of the meteorological variables in Table 1. In January, the interannual variability of average surface level temperature and absolute humidity decreased significantly (F test with $p < 0.05$) under the future climate. There were marginally significant decreases ($0.10 < p < 0.05$) in the interannual variability of average temperature and mixing height. While the interannual variability of the other meteorological variables in Table 1 tended to increase for July, none of the land cell average increases was statistically significant.

3.2. $PM_{2.5}$

3.2.1. January

[17] The changes in average January $PM_{2.5}$ concentrations are shown in Figure 2. Concentrations decreased over most of the domain and increased slightly in the Northeast, resulting in a net decrease of 0.3 $\mu g\ m^{-3}$ (−5%). The changes in average concentrations of the various PM

Table 2. Changes (Future – Present) in Land Cell Average $PM_{2.5}$ Species Concentrations in January and July[a]

Species	January	July
Total $PM_{2.5}$	−0.33 (−5%)	**+2.48 (42%)**
Sulfate	−0.17 (−8%)	**+1.37 (54%)**
Nitrate	−0.01 (−0%)	+0.03 (16%)
Ammonium	−0.04 (−4%)	**+0.50 (55%)**
SOA	−0.04 (−10%)	**+0.54 (38%)**
POA	−0.08 (−6%)	+0.03 (4%)

[a]Changes in bold were statistically significant with t test p values <0.05. Others were not statistically significant. Concentrations are in $\mu g\ m^{-3}$.

species are shown in Table 2. All species had similar changes, decreasing by less than 10%. Moreover, all species showed the same general response pattern shown by total $PM_{2.5}$ in Figure 2, indicating that the major meteorological effect on January $PM_{2.5}$ concentrations is a physical, rather than a chemical, one. Given the large increase in January precipitation in Table 1, changes in wet deposition due to changes in precipitation appear to be the dominant effect. The statistical significance of the changes in January $PM_{2.5}$ is shown in Figure 3. The main statistically significant changes were decreases in the Ohio Valley region. In nonsignificant areas, changes may simply be due to interannual variability.

[18] The effect of meteorological changes on U.S. 24-h $PM_{2.5}$ air quality standard exceedances was rather small in January. Under both present and future climates, the number of land grid cells exceeding the air quality standard (35 $\mu g\ m^{-3}$) was small, though there was a small increase due to changes in climate. Under present-day conditions 11 land grid cells exceeded the air quality standard in an average January, while under future conditions the average was 67 land grid cells (compared to over 5000 land grid cells in the modeling domain). The maximum number of cells exceeding the standard in any present-day January was 32, while the maximum for future climate Januaries was 83. The increases in January PM air quality standard exceedances primarily occurred in the Northeast, where average $PM_{2.5}$ concentrations increased because of meteorological changes (Table 3). These exceedance figures are quite small, however, compared to those of July (section 3.2.2).

[19] The regional responses of the various PM species are shown in Table 3. In January in a given region, nearly all species responses had the same sign. All species increased in the Northeast despite an increase in precipitation, indicating that other effects, such as humidity and wind speed, were important. PM concentrations decreased over most of the rest of the domain, while the increase in nitrate concentrations over the Northeast is possibly caused by increased absolute humidity (Table 1), which leads to increased partitioning of nitrate into the condensed phase [*Seinfeld and Pandis*, 2006]. Considering the lack of statistical significance of most changes in meteorology and in PM concentrations, though, it cannot be ruled out that these changes are simply a consequence of interannual variability.

[20] The regional average January $PM_{2.5}$ concentration and the interannual range of monthly average concentrations are shown in Figure 4. The range of monthly average concentrations decreased under future climate conditions in nearly all regions, possibly because of the decreased interannual variability of January climate variables (section 3.1). This smaller range of monthly average concentrations means that the interannual variability of January $PM_{2.5}$ concentrations decreased over much of the domain under future climate conditions.

[21] The effect of future climate changes on January $PM_{2.5}$ was dominated by the effects of changing precipitation. In a given location, all major species responded in the same direction, resulting in decreases in $PM_{2.5}$ over most of the domain and increases over the Northeast, where the average temperature increased and average wind speed decreased considerably. These results are largely dependent on the model-predicted 48% increase in January precipitation.

3.2.2. July

[22] The changes in average July $PM_{2.5}$ concentrations are shown in Figures 2 and 4. Increases in concentrations occurred over most of the domain, causing a land cell average change of +2.5 $\mu g\ m^{-3}$, or +42%. The changes in average species concentrations are shown in Table 2. In

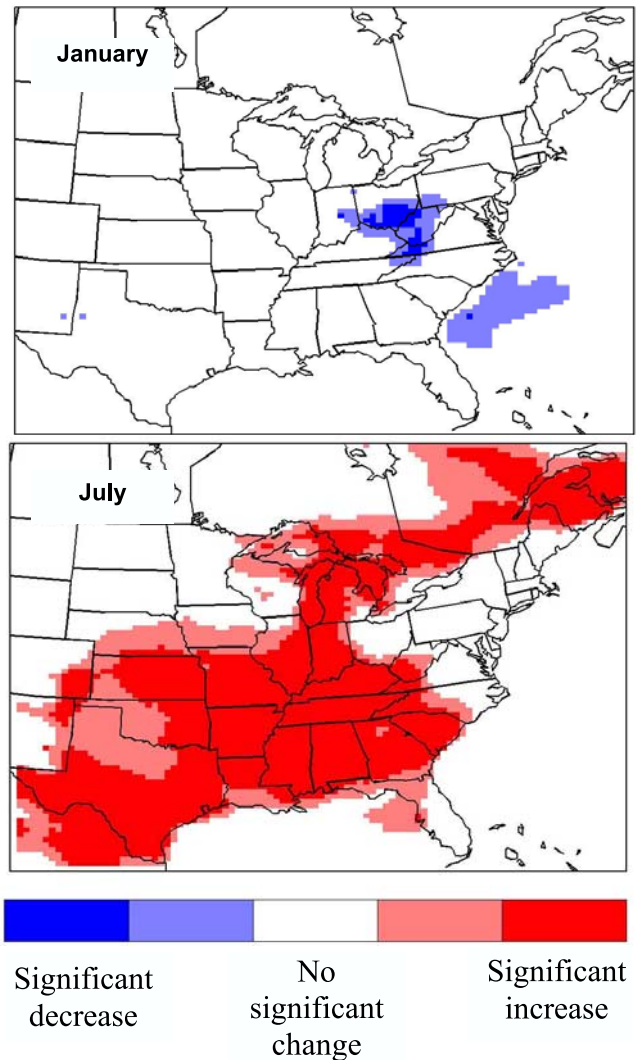

Figure 3. Map of significant changes in average (top) January and (bottom) July $PM_{2.5}$ concentrations. Areas with a "significant" change have t test p values <0.05. "No significant change" areas have p values >0.10. In intermediate areas, $0.10 < p < 0.05$.

Table 3. Regional Changes (Future – Present) in Average $PM_{2.5}$ Concentrations in the Five Regions[a]

Month	Region	Total $PM_{2.5}$	Sulfate	Nitrate	Ammonium	SOA	POA
January	plains	−0.38	−0.06	−0.16	−0.07	−0.03	−0.06
January	midwest	−0.16	−0.04	+0.04	+0.02	−0.03	*−0.15*
January	northeast	+0.35	+0.02	+0.18	+0.08	+0.04	+0.02
January	TX/OK	−0.61	−0.41	+0.05	−0.13	−0.07	−0.06
January	southeast	−0.74	−0.37	−0.09	−0.09	−0.09	−0.10
July	plains	+0.72	+0.43	+0.02	+0.19	+0.11	−0.04
July	midwest	**+2.94**	*+1.58*	+0.15	*+0.59*	**+0.49**	**+0.12**
July	northeast	+1.03	+0.35	−0.06	+0.15	+0.38	**+0.21**
July	TX/OK	**+3.34**	**+1.60**	+0.01	**+0.63**	**+1.10**	0.00
July	southeast	**+4.04**	**+2.67**	−0.03	**+0.86**	**+0.65**	*−0.11*

[a]Changes in italics were marginally statistically significant, with t test p values between 0.10 and 0.05, and changes in bold were statistically significant with t test p values <0.05. Others were not statistically significant. Concentrations are in $\mu g\ m^{-3}$.

July, the various PM species generally responded differently to meteorological changes, indicating complex chemical effects, unlike the simple effect of precipitation changes in January. The increases in average July $PM_{2.5}$ were statistically significant over much of the domain (Figure 3). Significant increases were generally located in the Southeast and Midwest. The interannual range of monthly average concentrations increased in four of the five regions of the domain and decreased in the Northeast (Figure 4). There were no significant changes in the interannual variability of the meteorological variables in Table 1 (section 3.1), however.

[23] Changes in sulfate concentrations were the largest of any species, in both absolute and relative terms (Table 2). Land cell average $PM_{2.5}$ sulfate increased by 1.4 $\mu g\ m^{-3}$, or 54%, under future climate conditions. The changes in sulfate concentrations are shown in Figure 5. Sulfate increased over nearly the entire domain, with some small decreases in parts of the Northeast. Most of the large increases in sulfate in Figure 5 were statistically significant. In the Northeast in Table 3, average sulfate concentrations increased while average nitrate concentrations decreased, likely indicating the effects of increased temperature on thermodynamic interactions between sulfate and nitrate (changing the temperature-dependent oxidation chemistry of SO_2, the amount of ammonia available, and the partitioning of semivolatile ammonium nitrate) [*Dawson et al.*, 2007b], and possibly changes in mixing height (Figure 1). Average concentrations of ammonium, sulfate, and SOA increased considerably under the changed climate (Table 2), though concentrations of nitrate and POA changed little. The increase in SOA concentrations indicates that chemical production effects are outweighing volatility effects. Since ammonium sulfate and SOA formation are dependent on temperature-dependent oxidation chemistry, this again shows the importance of temperature changes. Decreases in mixing height and wind speed and decreases in removal via wet deposition due to reduced precipitation (Table 1) increase concentrations of all PM species. Additionally, increases in humidity lead to increased partitioning of nitrate into the condensed phase [*Seinfeld and Pandis*, 2006]. Thus, it appears that changes in several meteorological parameters, especially mixing, precipitation, and temperature, have appreciable effects on July $PM_{2.5}$ concentrations. Interestingly, even though most of the changes in meteorological variables were not significant (Table 1), they led to changes in PM that were significant, indicating that subtle changes in climate may conceivably have important impacts on PM concentrations.

[24] In contrast to the increases in PM predicted over most of the domain in this study, *Tagaris et al.* [2007] predict decreases in all PM species in July, due largely to increased precipitation, resulting in a 10% decrease in total summer $PM_{2.5}$ over the United States. *Racherla and Adams* [2006], however, found increases of about 1 $\mu g\ m^{-3}$ in sulfate over the eastern United States in June and July, due primarily to decreased precipitation. The differences among these studies highlight the uncertainty of model predictions of some of the major drivers of PM concentrations, such as precipitation, wind speed, and mixing height.

[25] Changes in climate had a large effect on July exceedances of the 24-h $PM_{2.5}$ standard. Under present-day climate conditions, an average of 89 land grid cells exceeded a 24-h-average concentration of 35 $\mu g\ m^{-3}$ during an average July, compared to an average of 570 cells under future climate conditions. The present-day July with the highest number of land grid cells over the standard had 203 cells that exceeded the standard, while the maximum in a future

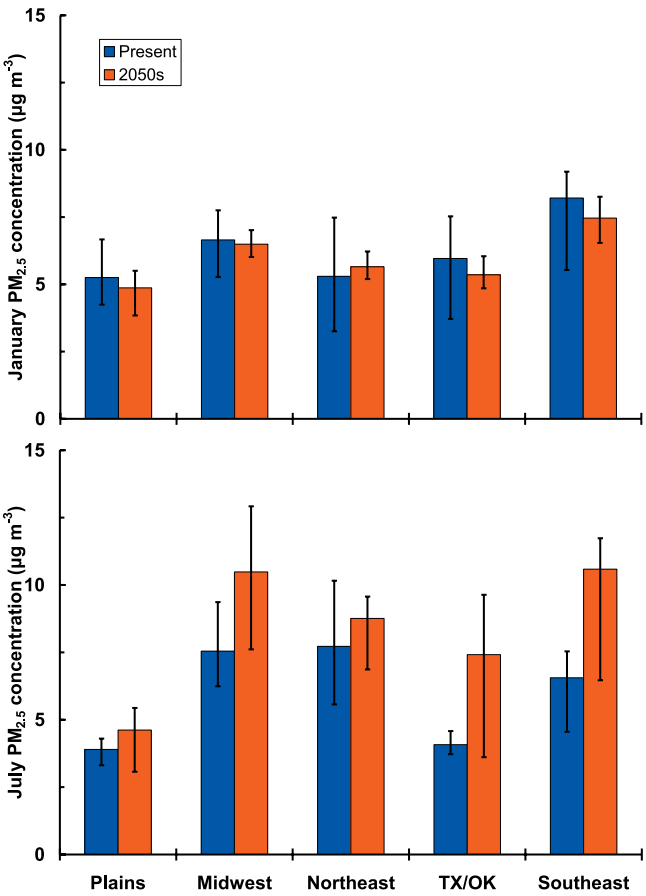

Figure 4. Average present and future climate (top) January and (bottom) July $PM_{2.5}$ concentrations in five regions of the modeling domain. Error bars show the full range of monthly average concentrations.

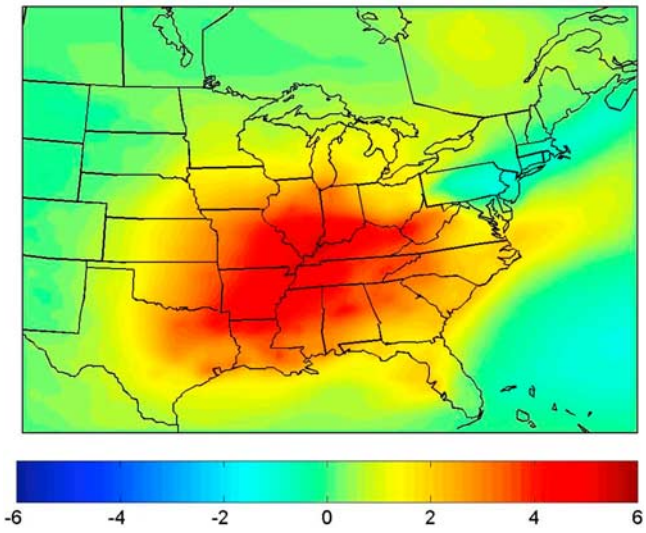

Figure 5. Changes (future – present) in average July $PM_{2.5}$ sulfate concentrations ($\mu g\ m^{-3}$).

July was 865. The number of Julys in which each grid cell exceeded the 24-h standard under present and future conditions is shown in Figure 6. The major increases were in the Midwest and Ohio Valley, where sulfate tends to be an especially important contributor to total $PM_{2.5}$. There were also appreciable increases in exceedances over the Southeast. The increases in temperature and decreases in wind speed and mixing height (Table 1) indicate that these increases in exceedances are likely due to decreased mixing and dilution and increased temperature under the future climate. *Mickley et al.* [2004] predicted an increase in stagnation in GCM simulations of the 2050s, resulting in an increase in pollution episodes.

[26] The changes in July $PM_{2.5}$ concentrations resulting from a changed climate were due to a mix of meteorological factors. Ventilation and temperature played important roles, and changes in precipitation and humidity appear to have also had effects. Average $PM_{2.5}$ concentrations increased over most of the domain as did exceedances of the 24-h air quality standard. $PM_{2.5}$ changes of this magnitude resulting from changes in climate could potentially have profound effects on public health and attainment of air quality standards.

3.3. Ozone

[27] The changes in average July MDA8 ozone concentrations are shown in Figure 7. The main effects of climate change were large ozone increases in the Southeast and smaller increases in the Midwest (Table 4). There were also small decreases over parts of the domain, especially the Northeast. The changes in MDA8 ozone were not statistically significant over most of the domain because of the large interannual variability in both present-day and future model predictions. These changes in ozone are due to a combination of the effects of the meteorological changes in Table 1. The large changes in mixing height and wind speed over the southern half of the domain are likely the main reasons for the increased ozone concentrations. In the southern areas in which ozone increased the most, average mixing heights decreased by 300–600 m, and wind speed decreased by 1–2 m s^{-1}. From *Dawson et al.* [2007a], changes in temperature and absolute humidity would also be expected to have effects on land cell average ozone concentrations, and these effects would be in opposition to one another. The effect of climate on land cell average MDA8 ozone was rather small: +1.7 ppb, though certain areas, such as Atlanta and Birmingham (Figure 7), had rather large effects (up to +15 ppb). The decreased ozone over much of the Northeast corresponds to a strip in which average mixing heights increased by up to 100 m.

[28] The regional changes in ozone and the interannual range of monthly average ozone are shown in Figure 8. The ozone increases over the Southeast and Midwest and the decreases over the Northeast were generally not statistically significant. These variabilities outweighed even the large changes in mean concentrations, causing the changes in means to be statistically insignificant. *Murazaki and Hess* [2006], in one of the only studies to examine statistical significance of ozone changes, did report significant

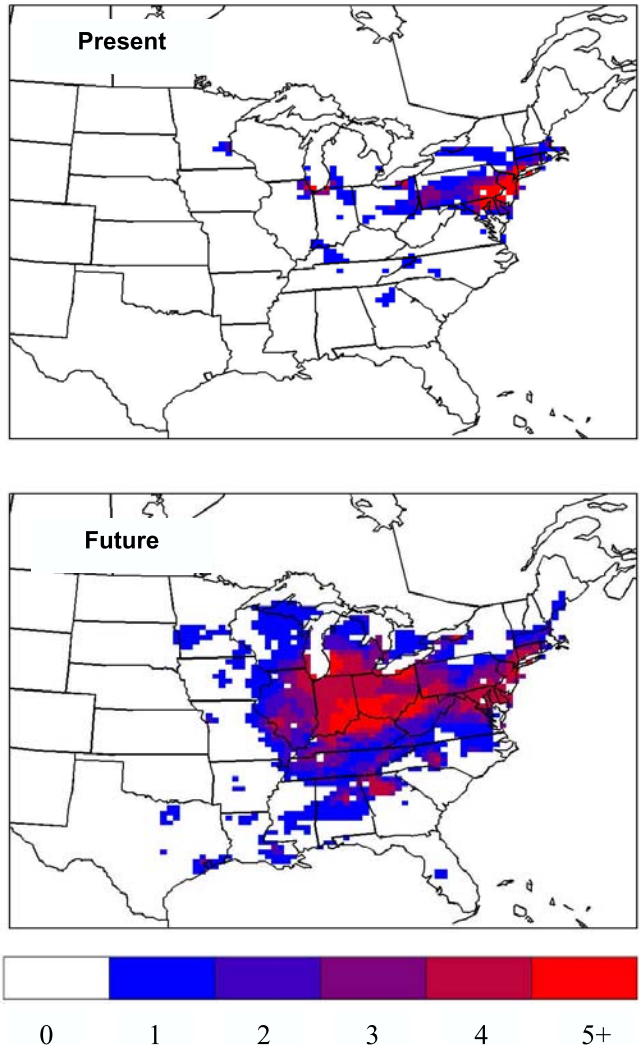

Figure 6. Map of number of Julys in which a land grid cell exceeds the $PM_{2.5}$ air quality standard of a 24-h-average concentration of 35 $\mu g\ m^{-3}$. (top) Six present climate and (bottom) five future climate Julys were simulated.

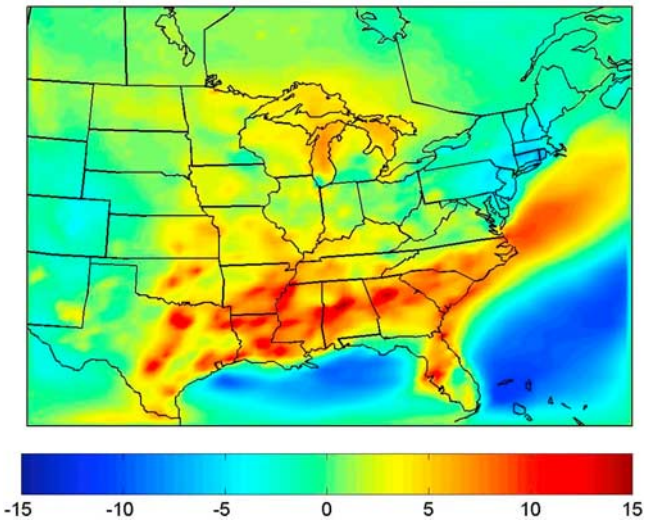

Figure 7. Changes (future – present) in July average MDA8 O_3 (ppb).

increases of ozone over much of the eastern United States; that study, however, represented both the present and future with 9-year ensembles and used the 2090s for the future time period, compared to 5- (or 6-) year ensembles and the 2050s, respectively, in the present study. Given the lack of information on statistical significance of ozone changes in other coupled modeling systems, it is currently impossible to determine whether this absence of statistical significance is unique to this study or if it is widespread.

[29] The increases in MDA8 ozone concentrations in the Midwest and near Atlanta were rather similar to those predicted by *Hogrefe et al.* [2004b], who predicted increases of 3–5 ppb over much of the Midwest due to climate and increases near 10 ppb in the Atlanta area. The effects predicted by GRE-CAPS over the rest of the Southeast, however, are much larger than those predicted by *Hogrefe et al.* [2004b]. *Racherla and Adams* [2006] predicted increases in annual average ozone concentrations of several ppb over much of the eastern United States, especially the Southeast. The changes in ozone predicted by *Tagaris et al.* [2007] were of similar magnitude, but included decreases in the Plains and Midwest and increases in the Northeast and Southeast, with a very small increase in the Southeast, which differs considerably from the large increases in ozone predicted in the Southeast in this study. *Wu et al.* [2008] predicted the largest increases in ozone over the Midwest and mid-Atlantic areas, with very minor changes over the Southeast. Most models seem to predict an increase in ozone of several ppb, therefore, over much of the eastern United States, though they disagree over the spatial pattern of ozone changes.

[30] In spite of the 1.7 ppb increase in land cell average MDA8 ozone concentrations, the average number of land grid cells exceeding the air quality standard decreased from 746 under present-day conditions to 618 under future climate conditions, a 17% decrease. The largest increases in MDA8 ozone occurred in areas that were usually over the standard under both present and future climate conditions (such as Atlanta). Therefore, these increases in ozone concentrations had little effect on the number of grid cells exceeding the air quality standard. *Murazaki and Hess* [2006] also calculated increases in ozone exceedances; in their study, the number of days per year with ozone greater than 80 ppb increased by up to 12 in areas of the eastern United States. *Nolte et al.* [2008] calculated increases in 95th percentile ozone concentrations over much of the eastern United States for the 2050s and the A1B scenario. Though these studies differ in scenarios and metrics from the present study, they all suggest an increase in severity or number of ozone episodes. The major decreases in exceedances of the air quality standard occurred over the Northeast, where average ozone concentrations decreased in the future climate simulations. This is likely due to the relatively high NO_x concentrations around the Northeast and to increasing mixing heights (Figure 1). The NO_2 from PAN decomposition resulting from increased temperature would decrease ozone concentrations in the high-NO_x Northeast, unlike most of the rest of the domain.

[31] The length of ozone episodes, however, largely increased under the future climate. The change in the amount of time with high ozone concentrations is shown in Figure 9. Large increases in the amount of time with high ozone concentrations occurred over much of the South, especially Louisiana and Texas. Similar lengthening of episodes was also predicted by *Mickley et al.* [2004] and *Murazaki and Hess* [2006]. Decreases in the amount of time

Table 4. Regional Changes (Future – Present) in Average MDA8 Ozone Concentrations due to Meteorological Changes in the Five Regions[a]

Region	Δ MDA8 O_3 (Climate) (ppb)	Δ MDA8 O_3 (Biogenics) (ppb)
Plains	0.5	0.3
Midwest	2.2	0.5
Northeast	−1.1	0.5
TX/OK	1.9	0.7
Southeast	4.5	1.5
Domain wide (land)	1.7	0.7

[a]No regional average change was statistically significant.

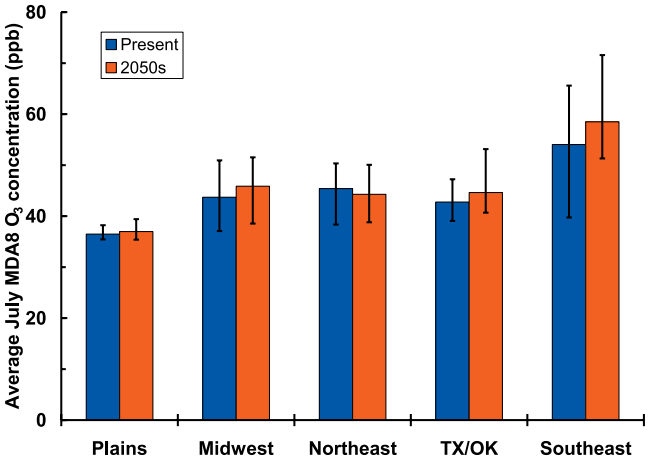

Figure 8. Average present and future climate July MDA8 ozone concentrations in five regions of the modeling domain. Error bars show the full range of monthly average concentrations.

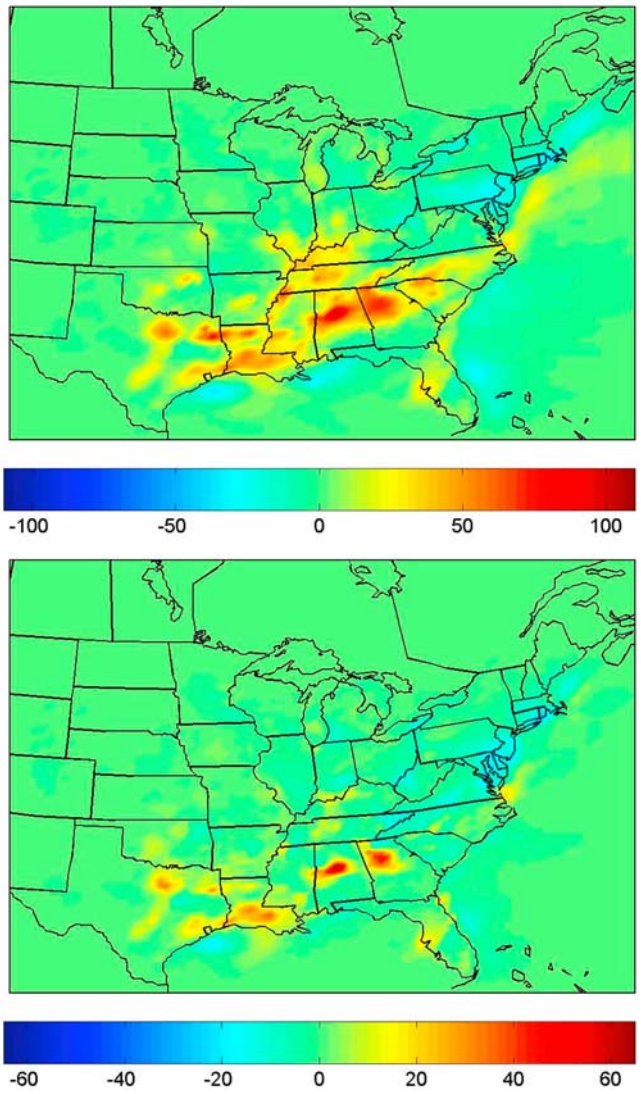

Figure 9. Difference in average number of July hours with ozone concentrations greater than (top) 70 ppb and (bottom) 84 ppb between future and present-day climates.

with high ozone concentrations occurred over much of the Northeast. The changes in the amount of time with ozone concentrations greater than 70 ppb were marginally significant in 60 grid cells, including 24 increases and 36 decreases, while the changes in amount of time over 84 ppb were significant in just 8 grid cells, split evenly between decreases and increases.

[32] The distributions of MDA8 ozone concentrations were rather similar between present and future climates. The probability density functions of MDA8 ozone concentrations for all present and future Julys are shown in Figure 10. The curves for the present years and future years lie very close to one another, indicating small changes in the distribution of ozone concentrations. Increased ozone concentrations due to climate effects, often referred to as the "climate penalty" [*Wu et al.*, 2008] could make the attainment of air quality standards more difficult in the Southeast or Midwest, especially if standards are lowered from their present value of 84 ppb to the proposed value of 75 ppb.

3.4. Effect of a 25% Increase in Biogenic Emissions

[33] Five separate simulations were run in which biogenic VOC emissions were increased by 25% under future climate July conditions. The effect of a 25% increase in biogenic emissions, roughly the amount predicted for the A2 scenario in the United States by *Racherla and Adams* [2006], on $PM_{2.5}$ concentrations during July was rather small. The land cell average increase in $PM_{2.5}$ concentration due to increased biogenic emissions was 0.12 μg m^{-3}. The changes in $PM_{2.5}$ concentrations due to increased biogenics are shown in Figure 11. These changes are primarily due to changes in biogenic SOA. Simulation average changes were less than 1 μg m^{-3} throughout the domain.

[34] A 25% increase in biogenic ozone precursors did generally increase ozone concentrations. The effect of this increase on ozone concentrations, with respect to the simulations in which the future climate was simulated with present-day biogenics, is shown in Figure 11. The largest effects were in the urban areas, because of their relatively high NO_x concentrations. The average change in MDA8 ozone due to the increase in biogenics was +0.7 ppb, though the increases were larger (up to 8 ppb) in southeastern cities, such as Atlanta, Houston, and Dallas. The change was positive in all five regions in Table 4. Even with both climate change and the 25% increase in biogenic emissions included in the future scenario, the changes in ozone concentration compared to the present-day were still generally not statistically significant.

[35] The effect of increased biogenics under a 2050s climate was similar to that predicted by *Hogrefe et al.* [2004b], who calculated increases of 2–6 ppb in MDA8 ozone over the Midwest. The effect over the Southeast was again larger in GRE-CAPS than in the modeling system of *Hogrefe et al.* [2004b]. The total net effect of climate and biogenics on MDA8 ozone compared to the present-day was an increase of 2.4 ppb, with an increase of over 20 ppb in some areas of the Southeast, especially urban areas such as Atlanta. The largest combined effects of climate and biogenics were over the Southeast across to Texas; there

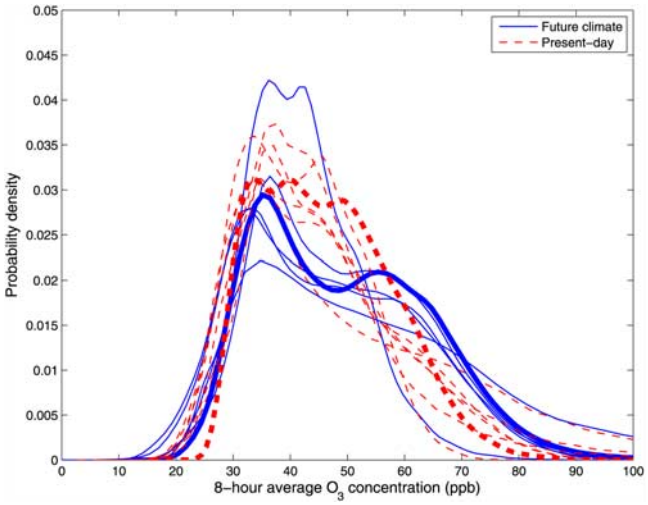

Figure 10. Probability density functions of MDA8 ozone concentrations for all modeled present and future Julys. Present and future averages are represented by bold lines.

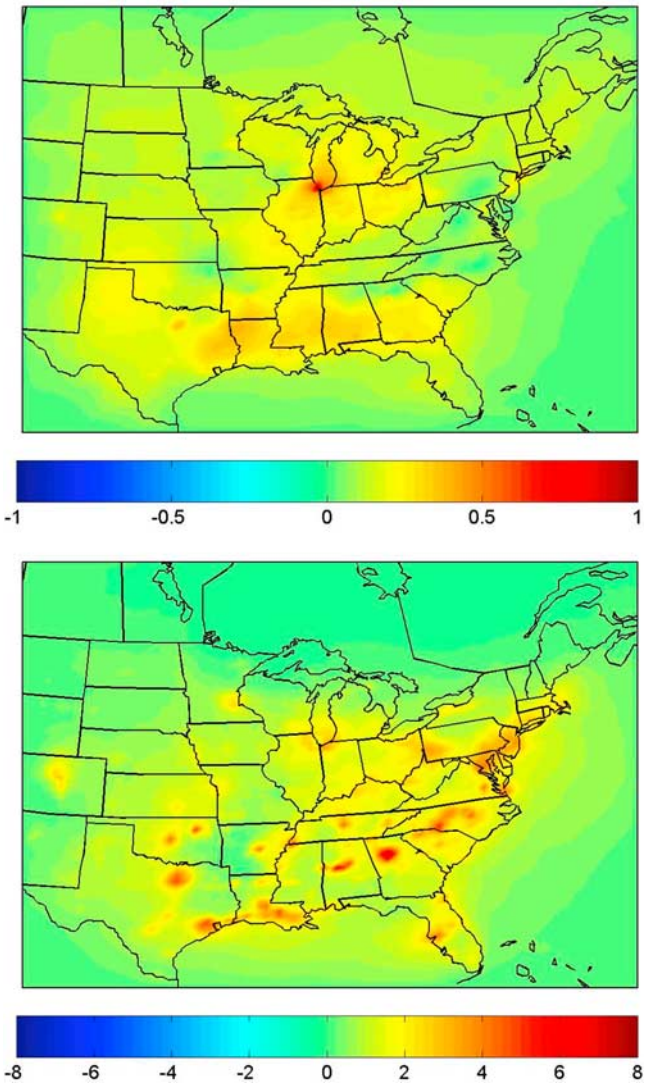

Figure 11. Changes in average (top) MDA8 ozone (ppb) and (bottom) $PM_{2.5}$ ($\mu g\ m^{-3}$) concentrations due to a 25% increase in biogenic emissions under future July climate conditions.

were also increases of several ppb in the Midwest, and slight decreases in the Northeast.

3.5. Effects of Individual Meteorological Parameters

[36] The changes in ozone and $PM_{2.5}$ concentrations predicted by GRE-CAPS were compared to those predicted by the meteorological sensitivities to perturbations in individual meteorological parameters from *Dawson et al.* [2007a, 2007b]. The summed PM sensitivities predicted the correct sign of the concentration change in both months. January $PM_{2.5}$ concentration changes were underestimated by 87% using the summed individual sensitivities, while July $PM_{2.5}$ changes were underestimated by 45%. However, the rather large errors indicate that summed individual sensitivities are not sufficiently rigorous to accurately predict the changes predicted by the full coupled modeling system. The changes in July MDA8 ozone concentrations using the linear meteorological sensitivities from *Dawson et al.* [2007a] and the land average meteorological changes equaled 0.0 ppb, compared to a GRE-CAPS-predicted value of 1.7 ppb. It appears that using domain average sensitivities and changes in meteorological is not sufficient for predicting changes in PM and ozone, especially considering that meteorological variables generally responded far from uniformly throughout the domain.

4. Conclusions

[37] The GRE-CAPS modeling system generally predicted a worsening of July air quality and improvement of January air quality between the present-day climate and the 2050s climate. Land cell average concentrations of $PM_{2.5}$ decreased by 0.3 $\mu g\ m^{-3}$ in January and increased by 2.5 $\mu g\ m^{-3}$ in July. January concentrations were most affected by changes in precipitation which increased under future climate conditions. Average July concentrations were affected largely by increases in sulfate. Decreases in mixing height and wind speed and increases in temperature led to the largest impacts on July $PM_{2.5}$ concentrations. The model-predicted increases in winter precipitation are predicted by most GCMs, indicating that there is some degree of robustness in at least the sign of the January responses to climate change. Average concentrations of $PM_{2.5}$ in the Philadelphia-Boston corridor of the Northeast had the opposite response from most of the rest of the domain in both January and July. While changes in January PM episodes were minor, July PM episodes increased dramatically under the future climate.

[38] Average MDA8 ozone concentrations increased during the summer over most of the eastern United States and especially in the Southeast, with a land cell average increase of 1.7 ppb. Separately, a 25% increase of biogenic VOC emissions led to an additional average increase of 0.7 ppb of ozone under future climate conditions, with the largest effect in urban areas in the South. Ozone episodes, as measured by exceedances of the 8-h-average air quality standard, became more prevalent as a result of climate change over much of the Southeast, though the total area over the ozone standard changed little between present and future climates. Most of the changes in monthly average MDA8 ozone concentrations were not statistically significant because of small ensemble size and large interannual variability, and neither were most of the changes in January $PM_{2.5}$ concentrations. Most of the increases in July $PM_{2.5}$ were significant, however. Additionally, representing the responses of ozone and PM to summed changes in individual meteorological parameters did not adequately capture the effects on changes in ozone and PM concentrations.

[39] Climate-related changes in ozone and particulate matter concentrations could have large effects on both public health and attainment of air quality standards in the future. ''Climate penalties'' of several ppb of ozone or several $\mu g\ m^{-3}$ of $PM_{2.5}$ could have effects on par with those of future emissions changes. Additionally, changes in interannual variability of ozone and PM concentrations could affect the setting and attainment of air quality standards. These climate-related changes in air quality will likely need to be taken into account in air quality planning, especially if air quality standards continue to be tightened. However, as climate models and chemical transport models are improved in the coming years, it can be expected that

estimates of the climate impacts on ozone and PM concentrations will be revised and, likely, improved.

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References

Adams, P. J., J. H. Seinfeld, and D. M. Koch (1999), Global concentrations of tropospheric sulfate, nitrate, and ammonium aerosol simulated in a general circulation model, *J. Geophys. Res.*, *104*, 13,791–13,823, doi:10.1029/1999JD900083.

Baertsch-Ritter, N., J. Keller, J. Dommen, and A. S. H. Prevot (2004), Effects of various meteorological conditions and spatial emission resolutions on the ozone concentration and ROG/NO_x limitation in the Milan area (I), *Atmos. Chem. Phys.*, *4*, 423–438.

Bernard, S. M., J. M. Samet, A. Grambsch, K. L. Ebi, and I. Romieu (2001), The potential impact of climate variability and change on air pollution-related health effects in the United States, *Environ. Health Perspect.*, *109*(suppl. 2), 199–209.

Byun, D. W., and J. K. S. Ching (Eds.) (1999), Science algorithms of the EPA Models-3 Community Multiscale Air Quality Model (CMAQ) modeling system, *Rep. EPA/600/R-99/030*, Off. of Res. and Dev., U.S. Environ. Prot. Agency, Washington, D. C.

Cess, R. D., et al. (1990), Intercomparison and interpretation of climate feedback processes in 19 atmospheric general circulation models, *J. Geophys. Res.*, *95*, 16,601–16,615, doi:10.1029/90JD01219.

Dawson, J. P., P. J. Adams, and S. N. Pandis (2007a), Sensitivity of ozone to summertime climate in the eastern USA: A modeling case study, *Atmos. Environ.*, *41*, 1494–1511, doi:10.1016/j.atmosenv.2006.10.033.

Dawson, J. P., P. J. Adams, and S. N. Pandis (2007b), Sensitivity of $PM_{2.5}$ to climate in the eastern US: A modeling case study, *Atmos. Chem. Phys.*, *7*, 4295–4309.

Dawson, J. P., P. N. Racherla, B. H. Lynn, P. J. Adams, and S. N. Pandis (2008), Simulating present-day and future air quality as climate changes: Model evaluation, *Atmos. Environ.*, *42*, 4551–4566, doi:10.1016/j.atmosenv.2008.01.058.

Gaydos, T. M., R. W. Pinder, B. Koo, K. M. Fahey, G. Yarwood, and S. N. Pandis (2007), Development and application of a three-dimensional aerosol chemical transport model, PMCAMx, *Atmos. Environ.*, *41*, 2594–2611, doi:10.1016/j.atmosenv.2006.11.034.

Gery, M. W., G. Z. Whitten, J. P. Killus, and M. C. Dodge (1989), A photochemical kinetics mechanism for urban and regional scale computer modeling, *J. Geophys. Res.*, *94*, 12925–12956, doi:10.1029/JD094iD10p12925.

Giorgi, F., and L. O. Mearns (2002), Calculation of average uncertainty range, and reliability of regional climate changes from AOGCM simulations via the "reliability ensemble averaging" (REA) method, *J. Clim.*, *15*, 1141–1158, doi:10.1175/1520-0442(2002)015<1141:COAURA>2.0.CO;2.

Grell, G. A., J. Dudhia, and D. Stauffer (1994), A description of the fifth-generation Penn State/NCAR Mesoscale Model (MM5), *NCAR Tech. Note, TN-398 + STR*, Natl. Cent. for Atmos. Res., Boulder, Colo.

Hansen, J., G. Russell, D. Rind, P. Stone, A. Lacis, S. Lebedeff, R. Ruedy, and L. Travis (1983), Efficient 3-dimensional global-models for climate studies: MODEL-I and MODEL-II, *Mon. Weather Rev.*, *111*, 609–662, doi:10.1175/1520-0493(1983)111<0609:ETDGMF>2.0.CO;2.

Heck, W. W., O. C. Taylor, R. Adams, G. Bingham, J. Miller, E. Preston, and L. Weinstein (1982), Assessment of crop loss from ozone, *J. Air Pollut. Control Assoc.*, *32*, 353–361.

Hogrefe, C., J. Biswas, B. Lynn, K. Civerolo, J.-Y. Ku, J. Rosenthal, C. Rosenzweig, R. Goldberg, and P. L. Kinney (2004a), Simulating regional-scale ozone climatology over the eastern United States: Model evaluation results, *Atmos. Environ.*, *38*, 2627–2638, doi:10.1016/j.atmosenv.2004.02.033.

Hogrefe, C., B. Lynn, K. Civerolo, J.-Y. Ku, J. Rosenthal, C. Rosenzweig, R. Goldberg, S. Gaffin, K. Knowlton, and P. L. Kinney (2004b), Simulating changes in regional air pollution over the eastern United States due to changes in global and regional climate and emissions, *J. Geophys. Res.*, *109*, D22301, doi:10.1029/2004JD004690.

Intergovernmental Panel on Climate Change (2000), *Special Report on Emissions Scenarios*, Cambridge Univ. Press, Cambridge, U. K.

Intergovernmental Panel on Climate Change (2001), *Third Assessment Report: Technical Summary of the Working Group I Report*, Cambridge Univ. Press, Cambridge, U. K.

Intergovernmental Panel on Climate Change (2007), Regional climate projections, in *Climate Change 2007: The Physical Science Basis*, chap. 11, pp. 887–892, Cambridge Univ. Press, Cambridge, U. K.

Karydis, V. A., A. P. Tsimpidi, and S. N. Pandis (2007), Evaluation of a three-dimensional chemical transport model (PMCAMx) in the eastern United States for all four seasons, *J. Geophys. Res.*, *112*, D14211, doi:10.1029/2006JD007890.

Lake Michigan Air Directors Consortium (2003), Base E modeling inventory, report, Midwest Reg. Plann. Organ., Rosemont, Ill. (Available at http://www.airtoxics.org/tech/emis/BaseE/baseEreport.pdf)

Leung, L. R., and W. I. Gustafson (2005), Potential regional climate change and implications to U.S. air quality, *Geophys. Res. Lett.*, *32*, L16711, doi:10.1029/2005GL022911.

Levy, J. I., T. J. Carrothers, J. T. Tuomisto, J. K. Hammitt, and J. S. Evans (2001), Assessing the public health benefits of reduced ozone concentrations, *Environ. Health Perspect.*, *109*, 1215–1226, doi:10.2307/3454743.

Liao, H., P. J. Adams, S. H. Chung, J. H. Seinfeld, L. J. Mickley, and D. J. Jacob (2003), Interactions between tropospheric chemistry and aerosols in a unified general circulation model, *J. Geophys. Res.*, *108*(D1), 4001, doi:10.1029/2001JD001260.

Mickley, L. J., P. P. Murti, D. J. Jacob, J. A. Logan, D. M. Koch, and D. Rind (1999), Radiative forcing from tropospheric ozone calculated with a unified chemistry-climate model, *J. Geophys. Res.*, *104*(D23), 30,153–30,172, doi:10.1029/1999JD900439.

Mickley, L. J., D. J. Jacob, B. D. Field, and D. Rind (2004), Effects of future climate change on regional air pollution episodes in the United States, *Geophys. Res. Lett.*, *31*, L24103, doi:10.1029/2004GL021216.

Murazaki, K., and P. Hess (2006), How does climate change contribute to surface ozone change over the United States?, *J. Geophys. Res.*, *111*, D05301, doi:10.1029/2005JD005873.

Nolte, C. G., A. B. Gilliland, C. Hogrefe, and L. J. Mickley (2008), Linking global to regional models to assess future climate impacts on surface ozone levels in the United States, *J. Geophys. Res.*, *113*, D14307, doi:10.1029/2007JD008497.

Racherla, P. N., and P. J. Adams (2006), Sensitivity of global tropospheric ozone and fine particulate matter concentrations to climate change, *J. Geophys. Res.*, *111*, D24103, doi:10.1029/2005JD006939.

Russell, G. L., J. R. Miller, and D. Rind (1995), A coupled atmosphere-ocean model for transient climate change studies, *Atmos. Ocean*, *33*, 683–730.

Schwartz, J., D. W. Dockery, and L. M. Neas (1996), Is daily mortality associated specifically with fine particles?, *J. Air Waste Manage. Assoc.*, *46*, 927–939.

Seinfeld, J. H., and S. N. Pandis (2006), *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*, 2nd ed., John Wiley, Hoboken, N. J.

Tagaris, E., K. Manomaiphiboon, K. J. Liao, L. R. Leung, J. H. Woo, S. He, P. Amar, and A. G. Russell (2007), Impacts of global climate change and emissions on regional ozone and fine particulate matter concentrations over the United States, *J. Geophys. Res.*, *112*, D14312, doi:10.1029/2006JD008262.

Wilkinson, J., and M. Janssen (2001), BIOME3, paper presented at National Emissions Inventory Workshop, U. S. Environ. Prot. Agency, Denver, Colo., 1–3 May. (Available at http://www.epa.gov/ttn/chief/conference/ei10/modeling/wilkenson.pdf)

Wu, S., L. J. Mickley, E. M. Leibensperger, D. J. Jacob, D. Rind, and D. G. Streets (2008), Effects of 2000–2050 global change on ozone air quality in the United States, *J. Geophys. Res.*, *113*, D06302, doi:10.1029/2007JD008917.

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